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Eco-efficiency evaluation of acetone-methanol separation processes using computational simulation

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ABSTRACT

The evaluation of environmental parameters of industrial processes plays a fundamental role in reducing not only their ecological impacts but also the costs associated with plant operations. In this regard, this work aims to evaluate, through eco-indicators and with the aid of computational simulation, the energy consumption, the CO_2 emissions and the water consumption of three different acetone-methanol separation plants, namely, Pressure Swing Distillation, Conventional Extractive Distillation and Energetically Integrated Extractive Distillation. Said processes were compared to one another in terms of their environmental efficiencies by means of the Eco-efficiency Comparison Index (ECI) method, which demonstrated that the integrated process is the most ecologically friendly.

1. Introduction

Notwithstanding the current concerns related to global warming, the world has been facing an alarming increase in greenhouse gases emissions, especially when it comes to industrial activities. In addition, the inappropriate raw materials exploitation, together with other environmental burdens such as the excessive consumption of water and energy, has compelled the companies and governments to seek alternatives for mitigating such ecological impacts.

The development of new technologies has become necessary to provide industrial processes with a better “eco-efficiency”, that is, the efficiency related to raw material consumption, product quality and environmental effects, in addition to the traditional financial income.

Eco-efficiency is a common object of industries that aim at sustainability, since it represents a direct relationship between economic development and its corresponding ecological impacts [1] and is based on the evaluation of the so-called “eco-indicators”. An eco-indicator is generally represented by the ratio of an environmental variable (e.g. energy consumption, CO_2 emissions, wastewater generation, water consumption, among others) to an economic variable (either net income or production rate) [2,3,12].

One of the main purposes of eco-indicators is to improve decision-making procedures by providing guidelines for assisting safe economic and environmental decisions. Eco-indicators also enable the prediction of the environmental impacts resulting from economic activities, which can be taken into consideration during the design phase of a new process, thus allowing the selection of more eco-efficient routes/configurations or improving an existing process.

In order to demonstrate the usefulness of eco-indicators as a convenient tool for predicting and comparing the eco-efficiency of industrial processes, three different acetone-methanol separation technologies were evaluated, namely, Pressure Swing Distillation (PSD), Conventional Extractive Distillation (CED) and Energetically Integrated Extractive Distillation (EIED). The analysis was carried out by quantifying their respective CO_2 emissions, as well as their water and energy consumption, with the aid of computational simulation.

2. Literature review

The increase in greenhouse gases emissions and the excessive

Abbreviations: CAPEX, capital expenditure; CED, conventional extractive distillation; CW, cooling water; CWP, cooling water pump; CWR, cooling water return; CWT, cooling water tower; ECI, eco-efficiency comparison index; EIED, energetically integrated extractive distillation; ESCAP, economic and social commission for Asia and the Pacific; FW, feed water; FW.R, feed water to reactor; FWP.B, feed water Pump to boiler; FWP.R, feed water pump to reactor; hps, high pressure steam; IPCC, Intergovernmental Panel on Climate Change; lps, low pressure steam; MCTI, Brazilian Ministry of Science, Technology and Innovation; mol, molar; mps, medium pressure steam; OPEX, operating expenditure; PSD, pressure swing distillation; UNIQUAC, universal quasi chemical; WBCSD, World Business Council for Sustainable Development

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Nomenclature

Symbols

l	Side of the triangle
n	Number of eco-indicator
$Q1$	Pump P-100 energy (MW)
$Qcond1$	Condenser E-101 energy (MW)
$Qcond2$	Condenser E-102 energy (MW)
$Qreb1$	Reboiler E-103 energy (MW)
$Qreb2$	Reboiler E-104 energy (MW)
S_a	Minor triangle area
S_T	Major triangle areaMajor triangle area
S_T*	Largest possible area

Greek Symbols

θ	Angle formed between two sides of the triangle

Sub- and superscripts

*	Largest possible
A	Side a
a	Minor area
B	Side B
i	Minor triangle
T	Major area

consumption of water and energy, along with other environmental burdens, have brought forth critical concerns related to the effects on the environment. Zhou and coworkers [7] stated that approximately 40% of the global CO_2 emission result from the processing industry, which is responsible for one third of the global energy use. This industry also stands out on water expending account, corresponding to approximately 23% of the global water consumption.

In view of such alarming scenario, the industries have been developing new studies and techniques associated with the economic and environmental performance of their activities in order to mitigate the resulting ecological impacts. In this regard, the concepts of eco-efficiency and eco-indicators play a crucial role because they can be utilized for evaluating the ecological performance of economic activities through a relationship between an environmental variable and an economic variable [4].

ESCAP (2009) [5] defined eco-efficiency as “a link between business and sustainable development”, being usually evaluated through the determination of eco-indicators and related to carrying out industrial operations in parallel to environmental preservation.

The analysis of eco-indicators provides information on the environmental performance of an industrial process that is related to its financial performance and improves the prediction of economic and ecological impacts [2]. Eco-indicators are generally defined as shown in Eq. (1). For evaluation purposes, the lower the value of the eco-indicator the more eco-efficient the process.

$$Eco - indicator = \frac{Environmental \quad Variable(unit)}{Economic \quad Variable(production \quad rate)} \tag{1}$$

Regarding industrial operations, the environmental variable may represent several categories such as energy consumption, CO_2 emissions, water consumption and wastewater generation, among others. The economic variable, in turn, is usually related to production rate [6–9]. However, when the objective is maximizing the economic variable, generally the eco-indicator is determined by the inverse ratio of Eq. (1) [10,40].

There are several other methodologies that have been applied to eco-efficiency analysis of chemical processes. Ecoindicator-99 [40], for instance, is a method which has been also applied for the sustainability evaluation of industrial processes. Hofstette et al. [40] applied Ecoindicator-99 for a life-cycle assessment (LCA) of two case studies: a waste solvent incineration plant and a batch distillation column. Such methodology was also applied by Luis et al. [41] in the analysis of batch and continuous distillation for the treatment of four waste-solvent mixtures typically produced in the chemical industry. Furthermore, five environmental indicators, namely Eco-indicator 99, UBP-97, global warming potential (GWP), cumulative energy demand and CO_2-balance, were applied by Amelio et al. [42] to analyze the production of solvents. The authors showed that said indicators led to the same conclusions for the evaluated mixtures, with some exceptions for UBP-97. Sánchez-Ramírez et al. [43], in turn, showed that eco-efficiency analysis can be applied to optimization studies of chemical plants. The authors have proposed the optimization of a biobutanol production plant by using the Ecoindicator-99 as one of the objective functions.

Eco-efficiency has also been widely studied in the literature for several industry fields such as iron [11], steel [6], petrochemical [10], separation of acetone-methanol [13], among others. Most studies regarding industrial eco-indicators relate to both energy consumption and CO_2 emissions, since the former is directly associated with the latter. This is due to both the highly perilous ecological impact resulting from greenhouse gases emissions and the connection between energy consumption and economic potential [14].

In the following sections we describe the main eco-indicators used in this study.

2.1. Eco-indicators

2.1.1. Energy consumption eco-indicator

Energy consumption is inextricably associated with ecological impacts since, in many cases, fossil fuel combustion is necessary to generate electricity and vapor energy, for example. Particularly in relation to industrial operations, the current energy demand from industrial processes is not only extremely high but also expected to grow. Therefore, process optimization techniques (e.g. thermal integration) have become a crucial strategy for reducing energy use and thus the consequent environmental impacts [15].

The joint evaluation of the energy consumption of a process and its economic performance can be carried out by determining the respective eco-indicator:

- Energy Consumption Eco-indicator – Ratio of the total energy (including electricity) used in a certain period to the total equivalent production rate (GJ/t).

2.1.2. CO_2 emissions eco-indicator

As previously stated, the CO_2 emissions eco-indicator is the most highlighted among the environmental indicators evaluated for industrial processes due to the notorious concern in relation to global warming. In fact, Adams [16] indicated that in the last decades there has been an increase in carbon dioxide emissions, especially from industrial activities. Such increase – approximately 500% between 1950 and 2012–corresponds to about 9.74 million tons of CO_2 emitted.

The joint evaluation of the CO_2 emissions of a process and its economic performance can be carried out by determining the respective eco-indicator:

- CO_2 Emissions Eco-indicator – Ratio of the total CO_2 emissions (combustion, indirect and fugitive) in a certain period to the total equivalent production rate (tCO_2/t).

2.1.3. Water consumption eco-indicator

The water consumption eco-indicator relates to the evaluation of the total amount of water (liquid and vapor) used in a process and may be analyzed for monitoring and control purposes when water is a feedstock. In order to determine such indicator, the refrigerating water and heating steam streams must be taken into account. It is also necessary to calculate the total amount of water needed to generate low pressure steam (lps), medium pressure steam (mps) and high pressure steam (hps) from the burning of natural gas in the boiler present in the utility plant. Such eco-indicator can be determined as follows:

- Water Consumption Eco-indicator – Ratio of the total water consumed in a certain period to the total equivalent production rate (m^3H_2O/t).

2.2. Eco-efficiency comparison

Understanding the ecological performance of industrial operations has become even more relevant for business environments due to the relationship between sustainability and economic development. Nevertheless, in many cases, single information are not sufficient to assist decision-making procedures such as, for instance, comparing different process routes and/or plant configuration. In view of this, industries have been developing techniques for better analyzing the eco-efficiency of their processes by means of a joint evaluation of eco-indicators.

The lack of eco-indicator calculation patterns generates a great challenge in terms of socio-environmental data consolidation. Thus, assembling a general analysis from a single indicator is an extremely arduous task. According to the WBCSD [17], this is also due to the divergence of priorities of each company and/or region. Therefore, searching for tools that enable the evaluation of several indicators at once can be an advantageous strategy.

Since environmental indicators can fall within different categories, methods that correlate different eco-indicators in a single analysis may assist in determining the global environmental impact of a process. An appropriate example of joint evaluation to be pointed out is the so-called “Eco-efficiency Analysis” developed by BASF [18] in 1996, which is based on the general evaluation of eco-efficiency through the estimation of numerous lifecycle assessment indicators.

Another method to be pointed out is the Eco-efficiency Comparison Index (ECI), developed by Pereira and coworkers [10]. The ECI method consists of grouping the normalized eco-indicators (calculated according to Eq. (1)) into radar-type charts and comparing them through a quantitative analysis regarding their respective resulting dimensionless area, as illustrated in Fig. 1. Thus, the ECI denotes an aggregated index aimed at processes monitoring according to their individual eco-efficiencies [10].

The ECI method aims to assemble a minimum limited area, which corresponds to a different approach when compared to other eco-efficiency comparison methods whose analysis is based on a “the bigger, the best” evaluation strategy. However, since such evaluation methods are unbounded, the comparison of indicator values may be troublesome when they tend to infinity. Furthermore, the ECI method is more plausible to be utilized by operators and engineers because it does not depend on the information on the financial income, which is usually not available for operational levels.

2.3. Computational simulation

Specialized companies have increasingly used computational modeling and simulation during the last decades due to the development of faster and more powerful hardware and software. Thus, the implementation of specific software for computational simulation has tightened the link between science and engineering [19].

Process simulation has become an advantageous strategy for the evaluation of process parameters and conditions in a relatively cheap basis. In fact, estimation of utilities costs and consumption, air emissions and waste generation are some examples of computer-aided environmental analysis already present in the literature [20].

2.4. Acetone-methanol separation processes

Acetone-methanol mixtures are mainly formed by solvent residues from the pharmaceutical industry [23]. Such mixtures are azeotropic, that is, liquid mixtures of substances that boils at a constant temperature lower or higher than of it components that retains the same composition in the vapor and liquid states [21]. In addition, since they are relevant for several industrial applications, the separation of such compounds is economically interesting.

The separation of acetone-methanol azeotropes can be performed through two principal techniques, namely, Pressure Swing distillation and Extractive distillation. The former is effective when the composition of the azeotrope varies significantly with pressure, while the latter is more convenient when it is possible to use an appropriate solvent [13].

Yeh et al. [22] were apparently the first ones to study the separation of acetone-methanol mixtures by extractive distillation. They found that either component may be obtained as overhead product depending on the solvent used. Modla and Lang [22] and Luyben [24], in turn, compared the annual costs associated with each technique, while Modla and Lang [13] evaluated the CO_2 emissions from two acetone-methanol separation extractive plants (conventional and thermally integrated) comprised of two batch rectifying columns. Nevertheless, such analysis was restricted to the estimation of CO_2 emissions only, while other environmental impacts such as water and energy consumption were not considered.

3. Methodology

3.1. Pressure swing distillation

Pressure Swing Distillation is commonly used when an azeotropic homogeneous mixture whose azeotropic composition is pressure sensitive is being processed, thus being a simple and economically feasible alternative to separate the mixture’s components [25].

Such technique makes use of two distillation columns operating at different pressures to alter the mixture’s relative volatility, thus enabling an effective separation. However, running two columns at different pressures results in a higher operational cost. Fig. 2 illustrates the example studied in this work, which was presented by Luyben [24].

In the process shown in Fig. 2, a feed stream comprised of 50 mol% acetone and 50 mol% methanol, at 540.0 kmol/h, is fed to a first

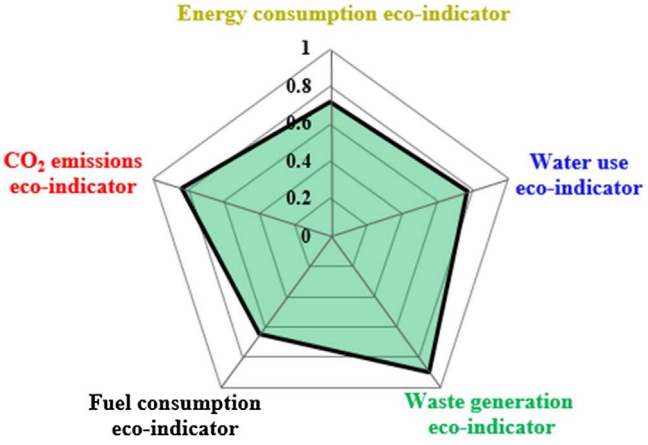

Fig. 1. Hypothetical radar-type chart correlating five eco-indicators.

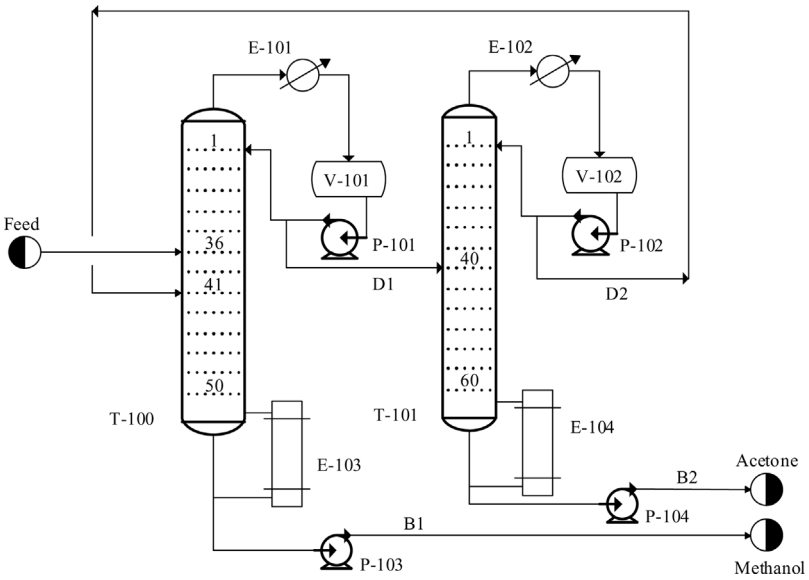

Fig. 2. PSD process' flow diagram.

distillation column T-100 that operates at 101.3 kPa, from which methanol is obtained as bottom product at 99.3 mol% and 269.7 kmol/h. T-100's distillate is sent to a second distillation column T-101 that operates at 1013 kPa, from which acetone is obtained as bottom product at 99.4 mol% and 270.3 kmol/h. T-101's distillate is recycled to the first column.

A more detailed description of this process, as well as the equipment parameters and stream conditions, are provided by Luyben [24].

3.2. Extractive distillation

Extractive distillation has been used for decades in bench scale and pilot plant facilities, while it has increasingly become significant to industrial scale processes [26]. In fact, several studies regarding this separation method are present in literature, namely, control strategies evaluation [24], phase equilibrium analysis [28,29] and solvent selection [22,24,27,30,31], among others.

Extractive distillation also makes use of two distillation columns and is based on the increase in the difference between the relative volatilities of the mixture's components through the addition of a third agent (i.e. a solvent). This agent and the most volatile compound(s) are removed at the bottom of the first column and separated in the second column. The non-extracted components are removed as distillate [26].

Comparing to PSD, Extractive distillation has a lower operational cost. However, since there is a need for adding a solvent into the system, it will be present as impurity in the final product, which represents a significant disadvantage. Figs. 3 and 4 illustrate the examples studied in this work, which were also presented by Luyben [24].

In the processes shown in Figs. 3 and 4, a feed stream comprised of 50 mol% acetone and 50 mol% methanol, at 540.0 kmol/h, is fed to a first distillation column T-100 that operates at 101.3 kPa, from which acetone is obtained as distillate at 99.5 mol% and 270.7 kmol/h. T-100's bottom product is sent to a second distillation column T-101 that operates at 101.3 kPa in the CED plant and 506.5 kPa in the EIED plant, from which methanol is obtained as distillate at 99.5 mol% and 271.1 kmol/h. T-101's bottom product is mixed with a make-up water stream prior to being recycled to the first column.

The thermal integration of the CED process consists in the energy exchange of T-101's top stage product against T-100's bottom product in a second reboiler E-103, as can be seen in Fig. 4. Subsequently, part of T-101's top stage product is retrieved as distillate while the rest returns to the column.

A more detailed description of both processes, as well as the equipment parameters, stream conditions and energy integration configuration, are provided by Luyben [24].

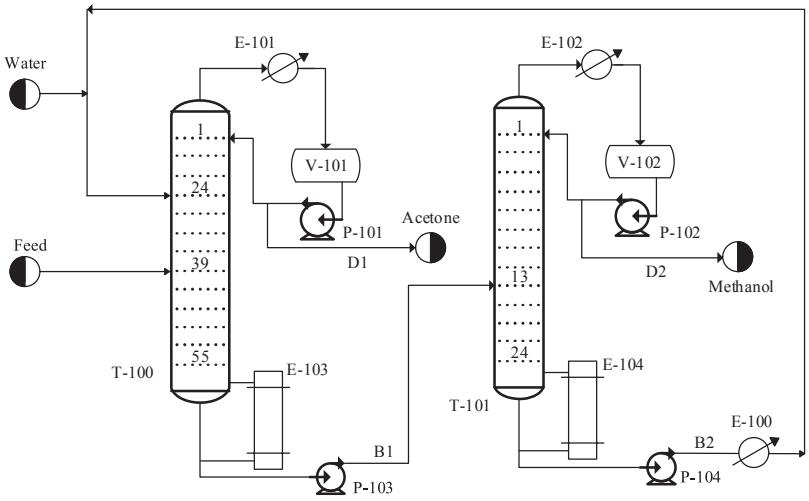

Fig. 3. CED process' flow diagram.

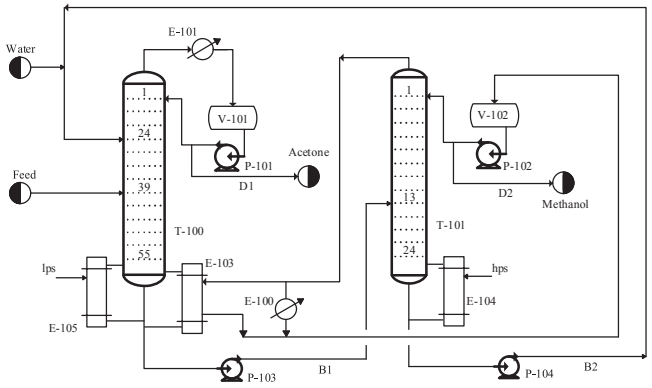

Fig. 4. EIED process' flow diagram.

3.3. Process simulation

The processes were simulated in Honeywell's UniSim® software by using the UNIQUAC (UNIversal QUAsiChemical) thermodynamic model, since it was also used by Luyben [21]. The plants simulated in this work are shown in Figs. 5–7, which illustrate each process in UniSim®'s environment. The flow rate and composition of the key component (top for PSD and botton for CED and EIED processes) were used as specification in distillation columns.

The energy integration configured in UniSim® was performed in a different configuration when compared to the one proposed by Luyben [24], which did not give any information either on the conditions of T-101's top stage product or cooler E-100's specifications, thus being not possible to reproduce such integration structure. In light of this hitch, to thermally integrate both columns, it was necessary to assign the energy stream of T-101's condenser to T-100's reboiler and then add a cooler E-101 before column T-101. The energy stream value of E-101 was set up to be equal to the energy required to bring stream B1's temperature to the desired inlet conditions.

3.4. Calculation of the eco-indicators

3.4.1. Energy consumption

The energy consumption eco-indicator was calculated by summing up all energy streams' requirements given by simulation and dividing the result by the total equivalent production rate of each process.

3.4.2. CO_2 emissions

In terms of CO_2 emissions calculation, the following definitions provided by the Intergovernmental Panel on Climate Change [32] were taken into consideration:

- Combustion Emissions – Burning of fossil fuels (liquid or gases) for heating or generating steam;
- Indirect Emissions – Associated with external steam/energy sources;
- Fugitive Emissions – Related to small leaks (valves, flanges, etc.), transport vehicles and especially burning off gases in the flare.

For the distillation processes studied, only combustion and indirect emissions were considered because emissions from leaks and transportation were deemed negligible and no flare system was present. If emissions from transport vehicles were taken into account, the final result would not be altered. Instead, only the absolute value would change, since the three plants yield approximately the same amount of acetone and methanol products. On the other hand, the estimation of leaks from valves and flanges depends on the valve type and is carried out via sampling of hydrocarbons in the actual plant. Therefore, the method used in this work is intended not only to approximate the value corresponding to the real emissions but especially to be used during the design phase in order to assist in decision-making procedures.

Considering that the present plants would be located in Brazil, the value of 0.0817 tCO_2/MWh must be used, since it corresponds to the annual average of CO_2 emissions related to electrical energy generation in Brazil in 2016. The conversion factors used in this work are presented in Table 1.

Both energy consumption and CO_2 emissions indicators were determined by considering that:

- Pumps operate with electrical energy and 75% efficiency (UniSim® default);
- Reboilers operate with steam generated by the combustion of natural gas in the boiler from the utility plant, which operates with 80% efficiency [34].

One should note that the procedure carried out in this work can be performed for any industrial process, regardless the site location. It is only necessary to utilize the corresponding conversions factors for each country/region.

3.4.3. Water consumption

To determine the water consumption eco-indicator, all cooling water and heating vapor streams used in the coolers, condensers and reboilers must be taken into account. Their respective flow rates were determined by simulating shell and tube heat exchangers.

It was also necessary to calculate the total amount of water required for generating low pressure steam (lps) and high pressure steam (hps) from the burning of natural gas in the boiler, as well as determine all water/steam losses in the utilities plant.

Since Luyben [24] did not provide the operating conditions for the water/energy utilities, it was necessary to resort to data and heuristics from literature. The utilities plant was divided into two main systems: cooling system and steam production system. The former is used for refrigerating and pumping cooling water to the coolers and condensers and is generally illustrated in Fig. 8 [35].

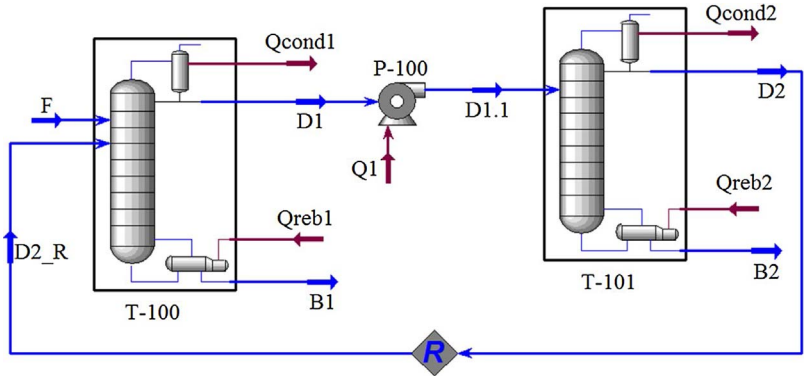

Fig. 5. PSD process simulated in UniSim®.

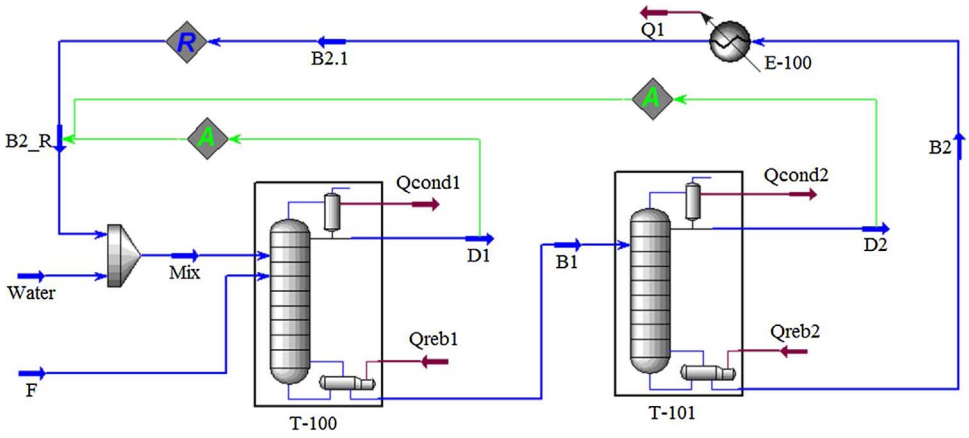

Fig. 6. CED process simulated in UniSim®.

In Fig. 8, CW, CWT, CWR and CWP stand for cooling water, cooling water tower, cooling water return and cooling water pump, respectively.

In relation to the steam production system, Seider et al. [36] stated that the water reaches the boiler at approximately 303.15 K and is heated until steam at the desired conditions (low or high pressure) is generated. Fig. 9 illustrates a generic steam production system.

In Fig. 9, FW, FWP.B, FWP.R and HPS stand for feed water, feed water pump to boiler, feed water pump to reactor (in case the process has a water-cooled reactor) and high pressure steam, respectively.

The heuristics for the operational conditions of the cooling and steam production plants were retrieved from literature [35,36] and are presented in Table 2.

Both utility systems were also simulated in UniSim® in order to facilitate the calculation of water/steam conditions (flow rate, temperature and pressure) required for each equipment, as well as their respective losses.

Table 1
Energy to CO_2 conversion factors.

Conversion data	Value	Reference
Electrical energy to CO_2	0.0817 tCO_2/MWh	IPCC [32]
Energy to CO_2 (Natural Gas basis)	0.20196 tCO_2/MWh	MCTI[33]

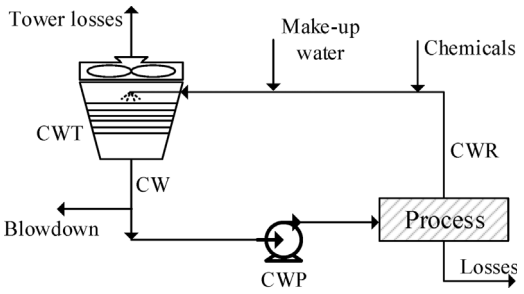

Fig. 8. Generic cooling system.

3.5. Eco-efficiency comparison index

The Eco-efficiency Comparison Index (ECI) was used to compare the eco-efficiencies of the three distillation plants studied in this work, with basis on the methodology developed by Pereira and coworkers [10].

Since each eco-indicator has a different unit, it was firstly necessary to normalize their values by grouping each indicator from the same category and dividing them by the highest value in that group. The respective index was normalized in the range from 0 (most eco-efficient) to 1 (least eco-efficient) in relation to that specific category[10]. The normalized values were then plotted in a radar-type chart, which formed a triangle since only three eco-indicator categories were considered in this paper.

The area of each minor triangle was calculated by using the Law of Sines, described in Eq. (2), and the resulting area of the major triangle is determined by summing up all minor triangle areas, as shown in Eq. (3):

$$S_a = \frac{l_A \cdot l_B}{2} \cdot sen\theta \tag{2}$$

$$S_T = \sum_{i=1}^{n} S_{ai} \tag{3}$$

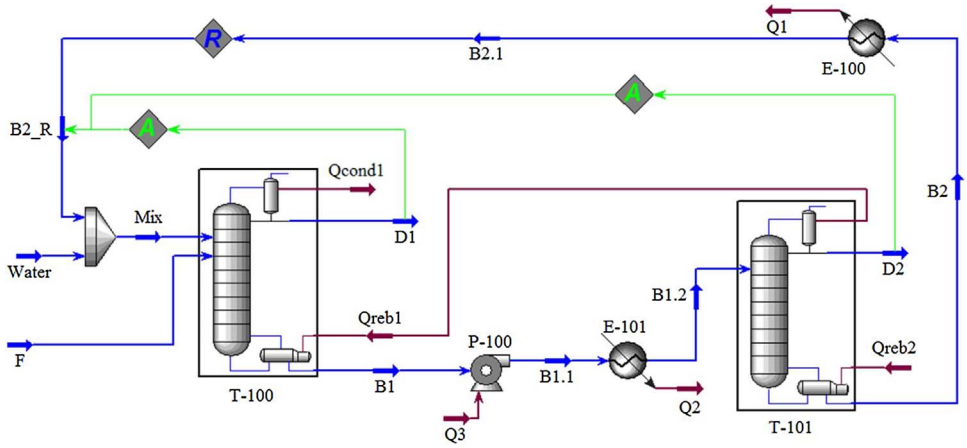

Fig. 7. EIED process simulated in UniSim®.

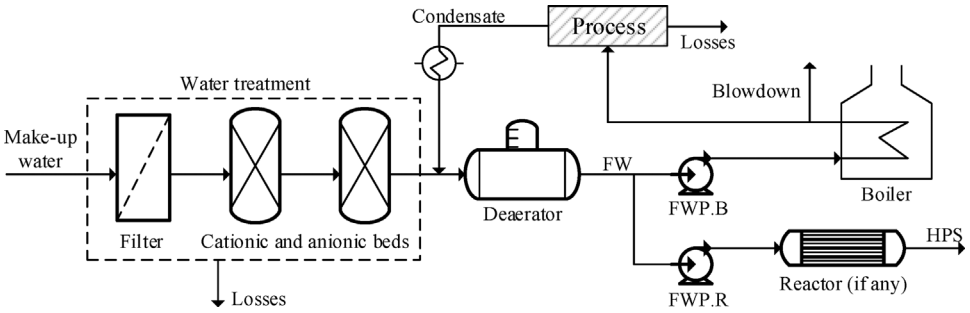

Fig. 9. Generic steam production system.

Table 2
Heuristics for utility systems − sent to and returning from process.

System	Temperature to – from, K	Pressure to – from, kPa
Cooling	301–318	516–496
Steam production (LPS)	408–407.7	310–310
Steam production (HPS)	527–526.8	4238–4238

Table 3
Simulation results for the PSD process.

	Stream	Temperature (K)	Pressure (kPa)	Flow rate (kmol/h)	Acetone	Methanol
F	Luyben [24]	320.0	101.3	540.0	0.5000	0.5000
	UniSim®					
D1	Luyben [24]	329.0	101.3	458.7	0.7500	0.2500
	UniSim®	328.5	101.3	457.2	0.7580	0.2420
	Error (%)	0.2	0.0	0.3	1.1	3.2
B1	Luyben [24]	345.0	101.3	269.7	0.0005	0.9995
	UniSim®	337.5	101.3	269.8	0.0007	0.9993
	Error (%)	2.2	0.0	0.0	20.0[a]	0.0
D2	Luyben [24]	337.0	101.3	188.4	0.4000	0.6000
	UniSim®	350.3	101.3	181.4	0.4161	0.5839
	Error (%)	3.9	0.0	3.7	4.0	2.7
B2	Luyben [24]	418.0	101.3	270.3	0.9940	0.0060
	UniSim®	416.8	101.3	270.3	0.9940	0.0060
	Error (%)	0.3	0.0	0.0	0.0	0.0

[a] Negligible since the error was found at the fourth decimal place.

$$\theta = 360°/n \tag{4}$$

One should note that θ is equivalent to 2π divided by the number of eco-indicators, since all axis are equidistant from one another and are set apart in a 360° circle.

In Eq. (2) S_a, l_A, l_B and θ refer to the area, side A, side B and the angle between two adjacent sides of the minor triangle, respectively.

In Eq. (3), in turn, S_{ai}, S_T and n correspond to the area of each minor triangle i calculated from Eq. (2), the area of the major polygon formed (in this case, a triangle) and the number of eco-indicators, respectively.

Finally, the ECI is calculated from Eq. (5), in which S_T is the area of the major triangle formed and S_T^* is the largest possible area to be calculated, which corresponds to the worst result (least eco-efficient).

$$ECI = \left(1 - \frac{S_T}{S_T^*}\right) \cdot 100\% \tag{5}$$

4. Results

The simulation of the three acetone-methanol distillation processes studied provided the required variables for calculating the energy consumption, CO_2 emissions and water consumption eco-indicators. Tables 3–5 present the stream results for the PSD, CED and EIED plants, respectively.

Some differences regarding streams conditions (e.g. temperature, pressure, etc.) were found when compared to the results obtained by Luyben [24], which may be due to the different software used by the author (Aspen Plus). Such differences are acceptable, since they were not higher than 5% of deviation (excepting those where the error was found at the fourth decimal place).

4.1. Energy consumption and CO_2 emissions

By summing up all energy stream values provided by simulation and following the IPCC [32] guidelines for converting energy use to carbon dioxide emissions, it was possible to determine the energy consumption and CO_2 emissions eco-indicators for each process. Table 6 shows the

Table 4
Simulation results for the CED process.

	Stream	Temperature (K)	Pressure (kPa)	Flow rate (kmol/h)	Methanol	Acetone	Water
F	Luyben [24]	320.0	101.3	540.00	0.5000	0.5000	0.0000
	UniSim®						
Water	Luyben [24]	320.0	101.3	1.73	0.0000	0.0000	1.0000
	UniSim®						
D1	Luyben [24]	329.0	101.3	270.70	0.9940	0.0011	0.0049
	UniSim®	329.2	101.3	270.71	0.9947	0.0010	0.0043
	Error (%)	0.1	0.0	0.0	0.1	9.1[a]	12.2[a]
B1	Luyben [24]	364.0	101.3	1369.00	0.0005	0.1974	0.8021
	UniSim®	359.1	101.3	1335.47	0.0006	0.2052	0.7942
	Error (%)	1.3	0.0	2.4	20.0[a]	4.0	1.0
D2	Luyben [24]	338.0	101.3	271.10	0.0025	0.9950	0.0025
	UniSim®	337.6	101.3	271.11	0.0027	0.9950	0.0023
	Error (%)	0.1	0.0	0.0	8.0[a]	0.0	8.0[a]
B2	Luyben [24]	378.0	101.3	1098.30	0.0000	0.0005	0.9995
	UniSim®	373.1	101.3	1084.36	0.0000	0.0003	0.9997
	Error (%)	1.0	0.0	1.3	0.0	40.0[a]	0.0

[a] Negligible since the error was found at the fourth decimal place.

Table 5
Simulation results for the EIED process.

	Stream	Temperature (K)	Pressure (kPa)	Flow rate (kmol/h)	Acetone	Methanol	Water
F	Luyben [24]	308.0	101.3	540.00	0.5000	0.5000	0.0000
	UniSim®						
Water	Luyben [24]	320.0	101.3	2.50	0.0000	0.0000	1.0000
	UniSim®						
D1	Luyben [24]	329.0	101.3	270.70	0.9950	0.0011	0.0039
	UniSim®	329.2	101.3	270.70	0.9950	0.0010	0.0040
	Error (%)	0.1	0.0	0.0	0.0	9.1[a]	2.6
B1	Luyben [24]	364.0	101.3	1369.00	0.0005	0.1970	0.8025
	UniSim®	354.7	101.3	1370.13	0.0005	0.1970	0.8025
	Error (%)	2.6	0.0	0.1	0.0	0.0	0.0
D2	Luyben [24]	385.0	506.5	270.50	0.0007	0.9950	0.0043
	UniSim®	384.9	506.5	271.10	0.0005	0.9948	0.0047
	Error (%)	0.0	0.0	0.2	28.6[a]	0.0	9.3[a]
B2	Luyben [24]	427.0	506.5	1097.40	0.0005	0.0005	0.9990
	UniSim®	425.5	506.5	1099.03	0.0003	0.0003	0.9994
	Error (%)	0.4	0.0	0.1	40.0[a]	40.0[a]	0.0

[a] Negligible since the error was found at the fourth decimal place.

Table 6
Energy consumption and CO_2 emissions eco-indicators.
[b]The value of 3.03 tCO_2/h was neglected because in the EIED process the reboiler E-103 operates with energy provided by T-101's condenser, as can be seen in Fig. 8. Therefore, E-103 should not be taken into account when calculating CO_2 emissions.

Plant	Source (simulation variable)	Emission type	Energy	Equipment Efficiency[a]	CO_2 emitted
	P-100 (*Q1*)	Indirect	0.011 MW	75%	0.001 tCO_2/h
	E-103 (*Qreb1*)	Combustion	14.42 MW	80%	3.64 tCO_2/h
	E-104 (*Qreb2*)	Combustion	6.82 MW	80%	1.73 tCO_2/h
PSD	Total		21.25 MW		5.37 tCO_2/h
	Production rate		24.34 t/h		24.34 t/h
	Energy eco-indicator		**0.87 MWh/t**		
	CO_2 eco-indicator				**0.22 tCO_2/t**
	P-100 (*Q1*)	Indirect	–	–	–
	E-103 (*Qreb1*)	Combustion	11.92 MW	80%	3.01 tCO_2/h
	E-104 (*Qreb2*)	Combustion	7.57 MW	80%	1.91 tCO_2/h
CED	Total		19.49 MW		4.92 tCO2/h
	Production rate		24.34 t/h		24.34 t/h
	Energy eco-indicator		**0.80 MWh/t**		
	CO_2 eco-indicator				**0.20 tCO_2/t**
	P-100 (*Q1*)	Indirect	0.005 MW	75%	0.001 tCO_2/h
	E-103 (*Qreb1*)	Combustion	11.99 MW[b]	80%	3.03 tCO_2/h[b]
	E-104 (*Qreb2*)	Combustion	16.02 MW	80%	4.04 tCO_2/h
EIED	Total		16.03 MW		4.04 tCO_2/h
	Production rate		24.34 t/h		24.34 t/h
	Energy eco-indicator		**0.66 MW h/t**		
	CO_2 eco-indicator				**0.17 tCO_2/t**

[a]The energy for the pumps was provided by UniSim® considering their efficiencies, which does not apply for the reboilers. Thus, the energy for the latter ones was corrected by dividing the values by the boiler efficiency.
[b]The value of 3.03 tCO_2/h was neglected because in the EIED process the reboiler E-103 operates with energy provided by T-101's condenser, as can be seen in Fig. 8. Therefore, E-103 should not be taken into account when calculating CO_2 emissions.

Table 7
Steam classification.

Steam	Temperature (K)	Pressure (kPa)
High pressure (hps)	504–527	2860–4238
Medium pressure (mps)	436–459	790–1135
Low pressure (lps)	394–408	210–310

results for all three distillation plants.

Although the product of the CED process has small concentrations of water, it demands less energy than the PSD plant. Thus, the amount of CO_2 emitted in the former is lower than in the latter. Moreover, by thermally integrating both separation columns of the CED process, one can note that the energy demand, and thus the CO_2 emission, can be severely decreased.

4.2. Water consumption

The amount of circulating water required by the coolers and condensers was determined from simulation by considering that the cooling water enters the equipment at 301.15 K (wet bulb temperature in Rio de Janeiro, Brazil) [37] and 516 kPa [35] and leaves at 318.15 K [36] and 496 kPa [35].

A similar procedure was carried out for calculating the flow rate of circulating steam in the reboilers. The temperature and pressure specifications for both inlet and outlet vapor streams were used as presented in Table 7 [36].

Turton and coworkers [35] recommended that the higher temperature values should be used and only latent heat should be considered. Thus, assuming that pressure loss is negligible, the steam exits the equipment as saturated liquid at the temperature of its respective steam class.

To calculate the water consumption eco-indicator it was necessary

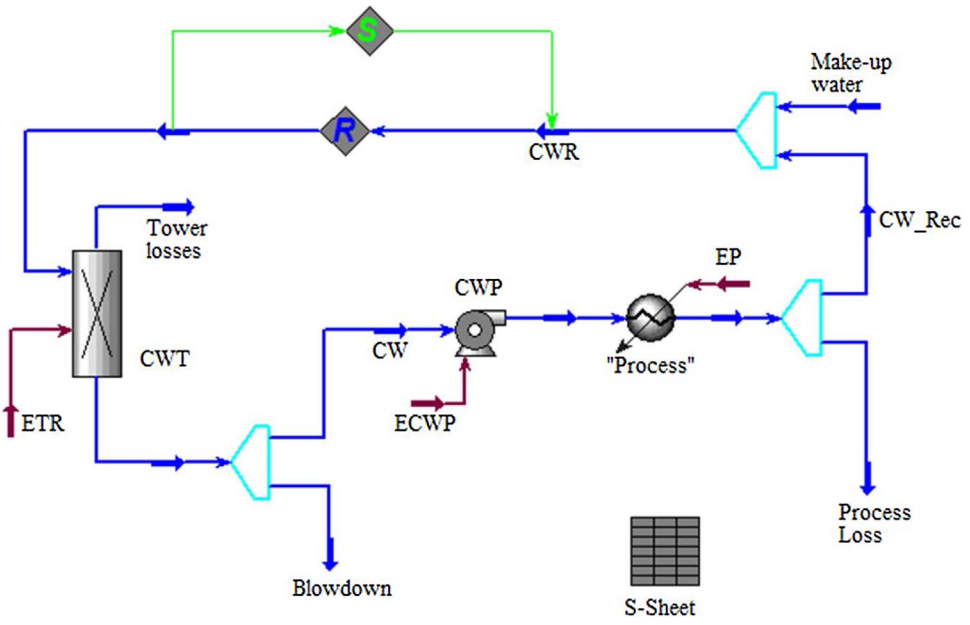

Fig. 10. Generic cooling system flow diagram simulated in UniSim®.

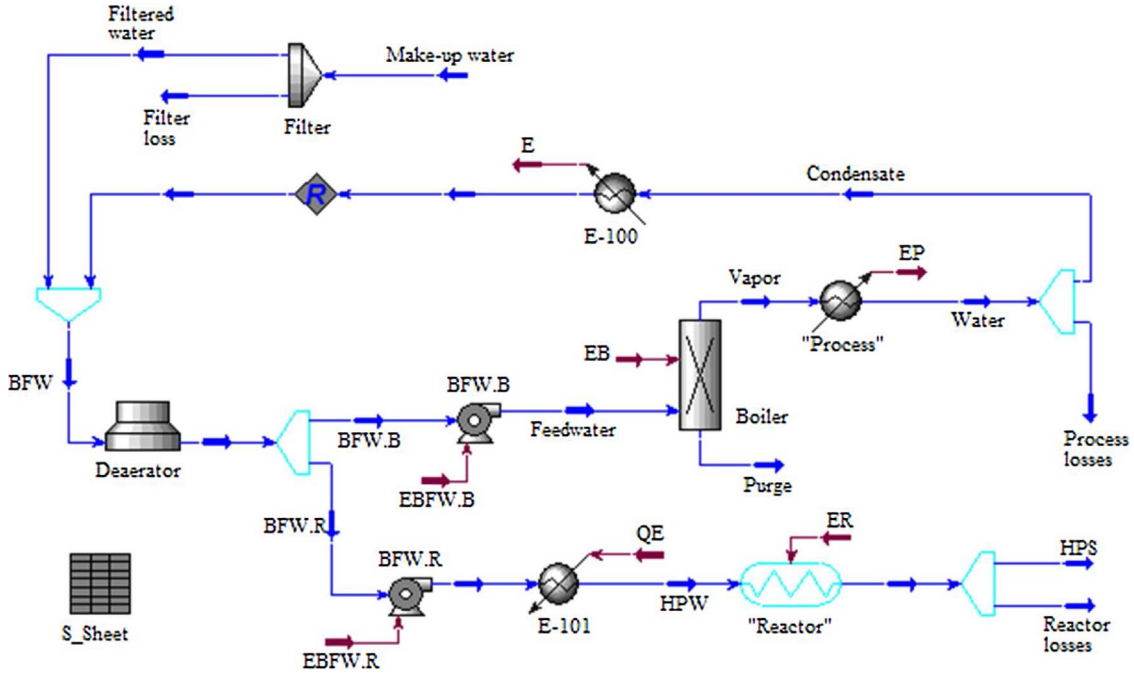

Fig. 11. Generic steam production system flow diagram simulated in UniSim®.

to account only for water losses in the cooling water and heating steam utility plants, which were simulated in UniSim® and are illustrated in Figs. 10 and 11, respectively. Both flow diagrams were based on the general representation of the cooling and steam production systems shown in Figs. 8 and 9.

Regarding the cooling water system, the losses in the tower (evaporative, windage, etc.), blowdown and losses in the process were set as 1%, 3% and 3%, respectively [38]. The spreadsheet "S-Sheet" then calculated, with basis on mass and energy balances, the amount of make-up water required to compensate the specified losses.

In relation to the steam generation system, in turn, the losses in the filter, purge and in the process were set as 2%, 3% and 20%, respectively [36,38]. Moreover, since none of the plants comprises a reactor, the flow rate of stream FW.R (Feed Water to Reactor) was set to zero. Then, it was necessary to set both the condensate return (80%) [36,39] and the boiler efficiency (80%) [34]. Finally, similarly to the cooling system, the spreadsheet "S_Sheet" calculated, the amount of make-up water required to compensate the specified losses.

Table 8 shows the results corresponding to the water consumption eco-indicator for each distillation process.

The water consumption of the EIED process is the lowest among all plants, which is due to the thermal integration of both columns that causes the energy demand to decrease significantly, thus resulting in a reduction of the utility requirements.

4.3. Eco-efficiency comparison index

With basis on their respective eco-indicators, the distillation plants were compared to one another to determine the least eco-efficient process. The two remaining plants were compared in order to obtain

Table 8
Water consumption eco-indicator results.

Plant	Water/Steam required in the process	Total losses in the utility plant (%)	Water/steam consumed (make-up)
	1225.4 m^3/h (water)	7.13	87.36 m^3/h
	44.8 m^3/h (steam)	20.61	9.24 m^3/h
PSD	Total		96.6 m^3/h
	Production rate		24.34 t/h
	Water use eco-indicator		3.97 m^3H_2O/t
	1173.7 m^3/h (water)	7.13	83.73 m^3/h
	41.3 m^3/h (steam)	20.53	8.48 m^3/h
CED	Total		92.21 m3/h
	Production rate		24.34 t/h
	Water use eco-indicator		3.79 m^3H_2O/t
	922.4 m^3/h (water)	7.14	65.81 m^3/h
	27.5 m^3/h (steam)	20.62	5.67 m^3/h
EIED	Total		71.48 m^3/h
	Production rate		24.34 t/h
	Water use eco-indicator		2.94 m^3H_2O/t

Table 9
Normalization of eco-indicators.

Eco-indicator	PSD	CED	EIED
Energy consumption	**0.87***	0.80	0.66
CO_2 emissions	**0.22***	0.20	0.17
Water consumption	**3.97***	3.79	2.94
Normalized eco-indicators			
Energy consumption	**1**	0.92	0.76
CO_2 emissions	**1**	0.91	0.77
Water consumption	**1**	0.96	0.74

* Highest values by category: normalization basis.

more detailed and complete data.

It was necessary to normalize the eco-indicators (i.e. dimensionless indicators) by dividing them by the highest value of the same category, as presented in Table 9.

Since the normalized indicators (from 0, most eco-efficient, to 1, least eco-efficient) are dimensionless, they could be jointly indexed. The results analyzed with basis on the ECI method are qualitatively illustrated in Fig. 12, which represents a radar-type chart.

The chart forms a triangle because only three eco-indicators were used. In fact, the number of sides of the resulting polygon is equal to the number of eco-indicators to be evaluated.

It can be verified that the ECI-formed triangle of the integrated plant is the smallest one, thus representing the best result (most eco-efficient). Hence, in order to quantitatively analyze such result, the area of each minor triangle was calculated according to Eq. (2).

The ECI was then determined from Eq. (5). Table 10 presents the results for all three processes.

Considering the same level of significance for each eco-indicator, the PSD process is approximately 14% and 42% less eco-efficient than the CED and EIED processes, respectively.

For a more detailed evaluation, the next step corresponds to the comparison between the CED and the EIED processes. In this regard, when a process is included in or excluded from the comparison analysis, the ECI must be recalculated because its results depend on the highest value of each category. Therefore, the same procedure was carried out for the extractive distillation plants, whose results are presented in Table 11.

Similarly, considering the same level of significance for each eco-indicator, the EIED process is 33.85% more eco-efficient than the CED process.

5. Conclusion

From the simulation of the three acetone-methanol separation processes studied, it was possible to obtain the required results for determining their environmental impacts through the evaluation of the energy consumption, CO_2 emissions and water consumption eco-indicators. Moreover, by using the Eco-efficiency Comparison Index method, all plants were compared to one another in terms of their eco-efficiency in order to determine which one is the most environmentally efficient.

By qualitative and quantitatively evaluating the eco-indicators for each process, it could be noted that the EIED plant presented the lowest environmental impact indices when compared to the PSD and CED plants. In fact, the EIED process is approximately 42% and 34% more ecologically efficient than the PSD and CED processes, respectively.

In light of such results, one can interpret energy integration as a crucial technique/solution for reducing utilities demand of a process, thus minimizing operational costs and environmental impacts. In addition, it could be verified the usefulness of eco-indicators in assisting in the comparison of different plant configurations, as well as the significance of computational simulation as a quick and low-cost technique for evaluating the economic and environmental parameters of processes.

In order to compare all three processes in a more detailed and complete manner, it would be interesting to develop future studies based on eco-indicators to analyze the feasibility of a thermal integration for the PSD process, as also suggested by Luyben [21], as well as

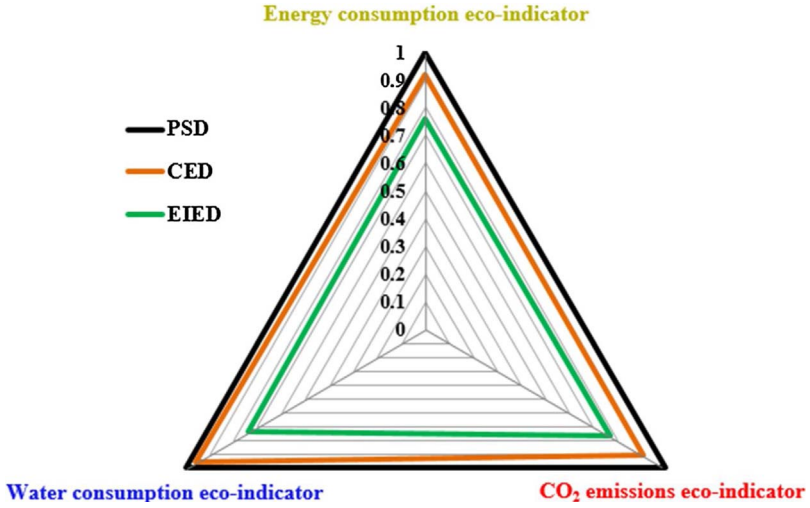

Fig. 12. ECI for all three processes.

Table 10
ECI for all three processes.

Eco-indicator × eco-indicator	PSD	CED	EIED
Energy consumption × water consumption	1	0.88	0.56
Water consumption × CO_2 emissions	1	0.87	0.57
CO_2 emissions × energy consumption	1	0.84	0.59
Sum	3	2.59	1.72
Triangle area (Eq. (2))	1.30	1.12	0.75
ECI (Eq. (5))	– –	**13.85**	**42.30**

Table 11
ECI for the extractive processes.

Eco-indicator × eco-indicator	CED	EIED
Energy consumption × water consumption	1	0.64
Water consumption × CO_2 emissions	1	0.65
CO_2 emissions × energy consumption	1	0.69
Sum	3	1.98
Triangle area (Eq. (2))	1.30	0.86
ECI (Eq. (5))	– –	**33.85**

determine the costs (i.e. CAPEX and OPEX) associated with each configuration.

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